

We claim :

1. A process for the oxidation of a C₂ to C₄ alkane to produce the corresponding alkene and carboxylic acid which process comprises contacting in an oxidation reaction zone, said alkane, molecular oxygen-containing gas, and the corresponding alkene and optionally, water, in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene, carboxylic acid and water, wherein in said process the molar ratio of alkene to carboxylic acid produced in said oxidation reaction zone is adjusted or maintained at a pre-determined value by controlling the concentrations of the alkene and optional water in said oxidation reaction zone and optionally by also controlling one or more of the pressure, temperature and residence time of the oxidation reaction zone.

2. An integrated process for the production of an alkyl carboxylate which process comprises the steps :

(a) contacting in an oxidation reaction zone a C₂ to C₄ alkane, a molecular oxygen-containing gas, the corresponding alkene and optionally, water in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene and carboxylic acid and water; and

(b) contacting in a second reaction zone at least a portion of each of said alkene and carboxylic acid produced in the first reaction zone, in the presence of at least one catalyst active for the production of alkyl carboxylate to produce said alkyl carboxylate, and in which process the molar ratio of alkene to carboxylic acid produced in said oxidation reaction zone is adjusted or maintained at a pre-determined value by controlling the concentrations of the alkene and optional water in said oxidation reaction

zone, and optionally by also controlling one or more of the pressure, temperature and residence time of the oxidation reaction zone.

3. An integrated process for the production of an alkenyl carboxylate which process comprises the steps:

- 5 (a) contacting in an oxidation reaction zone a C₂ to C₄ alkane, a molecular oxygen-containing gas, and the corresponding alkene and optionally, water in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene and carboxylic acid and water; and
- 10 (b) contacting in a second reaction zone at least a portion of each of said alkene and carboxylic acid produced in the first reaction zone and a molecular oxygen-containing gas, in the presence of at least one catalyst active for the production of alkenyl carboxylate to produce said alkenyl carboxylate,
- 15 and in which process the molar ratio of alkene to carboxylic acid produced in said oxidation reaction zone is adjusted or maintained at a pre-determined value by controlling the concentrations of the alkene and optional water in said oxidation reaction zone, and optionally by also controlling one or more of the pressure, temperature and residence time of the oxidation reaction zone;

20 4. A process as claimed in any one of the preceding claims which the molar ratio of alkene to carboxylic acid produced in the oxidation reaction zone is in the range 10 : 1 to 1 : 10.

5. A process as claimed in claim 4 in which the molar ratio of alkene to carboxylic acid produced in the oxidation reaction zone is in the range 0.8 : 1 to 1.4 : 1.

25 6. A process as claimed in any one of claims 2 to 3 in which alkene and/or carboxylic acid is separately recovered from the oxidation reaction product or separately added to the second reaction zone.

7. A process as claimed in any one of claims 1 to 3 in which the alkane is ethane, the corresponding alkene being ethylene and the corresponding carboxylic acid being acetic acid.

30 8. A process according to claim 2 wherein the alkyl carboxylate is ethyl acetate

9. A process according to claim 3 wherein the alkenyl carboxylate is vinyl acetate

10. A process according to claim 8 or claim 9 and wherein the molar ratio of alkene to carboxylic acid produced in the oxidation reaction zone is in the range 0.8 : 1 to

1.4 : 1.

11. A process according to any one of claims 1 to 3 wherein the concentration of alkene fed to the oxidation reaction zone is from greater than 0 and up to and including 50 mol% of the total feed, including recycles.

5 12. A process according to any one of claims 1 to 3 wherein the concentration of water fed to the oxidation reaction zone is from 0 to 50 mol% inclusive of the total feed, including recycles.

13. A process according to any one of claims 1 to 3 wherein alkene and water are fed into the oxidation reaction zone.

10 14. A process according to any one of claims 1 to 3 wherein the alkene and water are fed into the oxidation reaction zone in an alkene : water ratio of 1 to 0.1-250 by weight.

15. A process according to claim 14 wherein the ratio of alkene : water is 1 to 0.1-10 by weight.

15 16. A process according to claim 15 wherein the molar ratio of alkene : carboxylic acid is in the range 0.8 : 1 to 1.4 : 1.

17. A process according to any one of claims 1 to 3 in which the at least one catalyst in the oxidation reaction zone comprises molybdenum.

18. A process according to claim 3 in which the at least one catalyst in the second reaction zone comprises palladium.

19. A process according to any one of claims 1 to 3 in which the oxidation reaction is carried out at a temperature in the range 100 to 400 °C

20. A process according to any one of claims 1 to 3 in which the oxidation reaction is carried out at atmospheric or superatmospheric pressure

25 21. A process according to any one of claims 1 to 3 in which the oxidation reaction is carried out at a GHSV of 500-10,000 hr⁻¹

22. A process according to any one of claims 1 to 3 wherein the product stream from the oxidation reaction zone also comprises carbon oxides in an amount of less than 15 mol%.

30 23. A process according to any one of claims 1 to 3 in which the alkane is ethane, the corresponding alkene is ethylene, the corresponding carboxylic acid is acetic acid and wherein ethylene and water are fed into the oxidation reaction zone in a ratio of 1 to 0.1-10 by weight, the molar ratio of ethylene to acetic acid produced is in the range 0.8 :

1 to 1.4 : 1 and the product stream from the oxidation reaction zone also comprises carbon oxides in an amount of less than 15 mol%.

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